

Rheological Chaos of Frictional Grains

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A two-dimensional dense fluid of frictional grains is shown to exhibit time-chaotic, spatially heterogeneous flow in a range of stress values, σ , chosen in the unstable region of s-shaped flow curves. Stress controlled simulations reveal a phase diagram with reentrant stationary flow for small and large stress σ . In between no steady flow state can be reached, instead the system either jams or displays time dependent heterogeneous strain rates $\dot{\gamma}(\mathbf{r}, t)$. The results of simulations are in agreement with the stability analysis of a simple hydrodynamic model, coupling stress and microstructure which we tentatively associate with the frictional contact network.

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Discontinuous shear thickening is a ubiquitous phenomenon, observed in many dense suspensions [1, 2]. Simulations for non-Brownian particle suspensions [3–5] as well as for frictional granular media [6, 7] have highlighted the particular role played by frictional particle interactions. Discontinuous shear thickening implies a region of shear stress which (at finite Reynolds number, Ref. [8]) is not accessible to a homogeneous system in stationary state. What happens, if the system is forced into this regime by prescribing the stress at the boundary in the unstable region? One possibility is vorticity banding [9], corresponding to bands with different stress values at the same shear rate. However there is no clear evidence for persistent vorticity banding in experiment so far. Furthermore objections have been raised as to the possibility of vorticity banding as a stationary state: The pressure - in contrast to the shear stress - has to be the same across the interface of the bands; otherwise particle migration is expected to occur and thereby destabilise the interface. If stationary states are not accessible to the system, we expect to observe time-dependent, inhomogeneous states, either oscillatory or chaotic [10].

In this paper we show that spatio-temporal chaos occurs in a two-dimensional system of frictional granular particles, subject to an applied stress which is chosen in the unstable region of the flow curve. We present results from simulations and formulate a hydrodynamic model to derive a phase diagram and identify the regions of parameter space, where time-chaotic, inhomogeneous solutions are to be found.

We simulate a two-dimensional system of N soft, frictional particles in a square box of linear dimension L as detailed in [7]. The particles all have the same mass $m = 1$, but are polydisperse in size with diameters 0.7, 0.8, 0.9 and 1 in equal amounts. Normal and tangential forces, $\mathbf{f}^{(n)}$ and $\mathbf{f}^{(t)}$, are modeled with linear spring-dashpots of unit strength for both, elastic as well as viscous contributions. Thereby units of time, length and mass have been fixed [11]. Flow curves for other visco-elastic parameters are presented in the appendix. Coulomb friction is implemented with friction param-

eter $\mu = 2$, corresponding to the high friction limit. We expect the same qualitative findings as presented in this Rapid Communication for all values of $\mu > 0$ and refer to a systematic study of the μ -dependence in ref. [6]. In the stress-controlled simulations, a boundary layer of particles is frozen and the boundary at the top is moved with a force $\sigma L \hat{e}_x$, whereas the bottom plate remains at rest. In shear direction we use periodic boundary conditions.

Constitutive equation - Previous work [6, 7] has revealed discontinuous shear thickening for a range of packing fractions close to the jamming transition. The flow curves for frictional granular particles are well represented by the following constitutive equation

$$\dot{\gamma}(\sigma) = a\sigma^{1/2} - b\sigma + c\sigma^2, \quad (1)$$

where the term $-b\sigma$ is due to the frictional interactions and dependence on the packing fraction, ϕ , is implemented with $a = a(\phi)$ [7, 18]. This gives rise to the phenomenology of the van-der-Waals theory for a first-order phase transition: jamming first occurs at the critical point $\phi_c \cong 0.795$ which also marks the onset of hysteresis. A finite yield stress first appears at $\phi_\sigma \cong 0.8003$, while the generalized viscosity, $\eta = \sigma/\dot{\gamma}^2$, diverges only at $\phi_\eta \cong 0.819$. A similar, but not identical, sequence of characteristic packing fractions has been proposed in [6, 13]. Most remarkably is the existence (due to the b -dependent term) of a regime of shear stress which is unstable $\frac{\partial \dot{\gamma}}{\partial \sigma} < 0$, corresponding to s-shaped flow curves. In strain-controlled conditions such an s-shape leads to discontinuous shear thickening. In the van-der-Waals theory it corresponds to the coexistence region.

Simulations in the unstable regime - These s-shaped flow curves are indeed observed in the simulations (see Fig. 1) however only as transients and only in rather small simulation cells ($N < 24000$). In larger systems the s-shape is slowly eroded and vanishes completely above a certain system size. Instead a regime of continuous shear thickening develops. Closer inspection of the simulations in this regime reveals that no simple steady-state is reached. Rather, the system displays time-chaotic and spatially inhomogeneous behavior. An

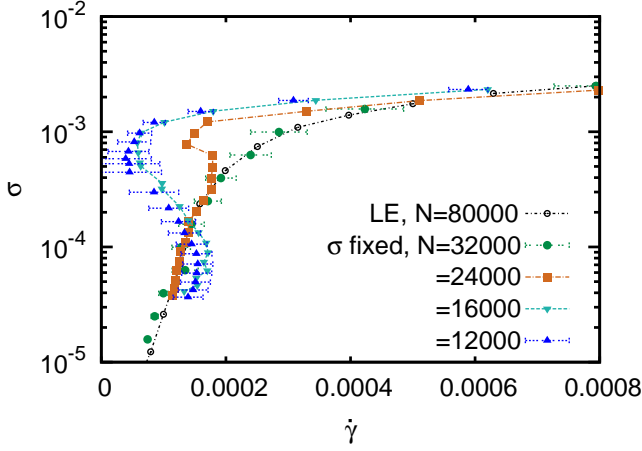
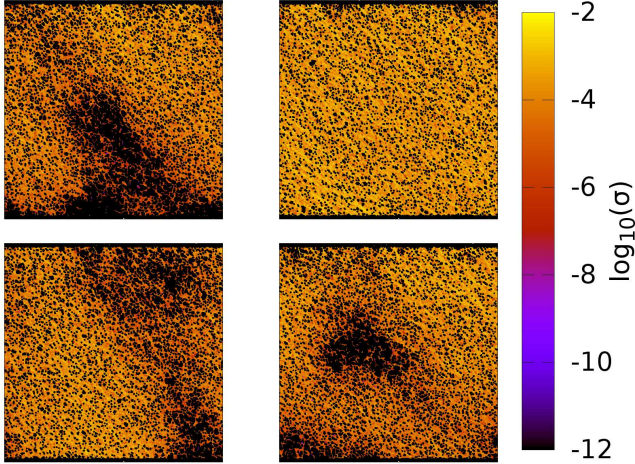


FIG. 1. Evolution of flow curves $\sigma(\dot{\gamma})$ with system size N . For small systems a pronounced s-shape is visible in transients before the system jams. In large systems no steady flow occurs; average over the time-dependent flow results in continuous shear thickening (stress controlled except for $N = 80.000$).



Video 1. Four snapshots of the local stress, revealing large scale, time-dependent structures; $\phi = 0.8035$.

example is shown in Video 1, which is a sequence of 4 snapshots of the local-stress field (movies are given in the supplementary material [19]). The system does not settle into a time-independent steady state on the timescale of the simulations. Instead one observes time-dependent large scale structures, e.g. shear bands which seem to propagate in the principal stress direction, alternating with approximately homogeneous states and random large scale structures. In Fig. 4 we show the corresponding time-dependent strain rate. One clearly observes irregular time-dependence with intermittent oscillatory periods.

Hydrodynamic model - In order to understand these time-dependent solutions and locate the regions of

parameter space, where they can occur, we now formulate a hydrodynamic model, determine its stationary states and analyse their stability. Our starting point is the momentum conservation equation in the form $\partial_t v_x = \partial_y \sigma_{xy}$. For simplicity we only consider a one-dimensional model, allowing for a velocity v_x in the flow direction, dependent on y only. In addition we introduce a variable $w(y, t)$ for the internal state or the microstructure of the fluid. Such a variable has been introduced for many complex fluids, such as liquid crystals, entangled polymer solutions and colloidal systems [9]. Here we associate it with the frictional contact network. In the simplest model we only consider a scalar variable, representing e.g. the number of frictional contacts [8, 15], but are aware that a tensorial quantity, such as the fabric tensor, might be more appropriate. We assume that the microstructure variable relaxes to a stationary state $\partial_t w = (w - w^*)/\tau$ which should vanish in the absence of stress $w^*(\sigma \rightarrow 0) \rightarrow 0$. Furthermore dynamic rearrangements occur only due to driving, so that $\tau^{-1} \propto \dot{\gamma}$. So far the model is the same as considered by Nakanishi et al. [14] for dilatant fluids. However, the coupling of stress and microstructure is different for the frictional grains under consideration. The frictional contacts reduce the flow and hence the velocity gradient. Starting from the constitutive equation for the strain rate of frictionless grains $\dot{\gamma}_0 = a\sigma^{1/2} + c\sigma^2$, we take the strain rate of our system of particles with friction to be $\dot{\gamma} = \dot{\gamma}_0 - w$. This completes the definition of the hydrodynamic model

$$\begin{aligned} \partial_t \dot{\gamma} &= \partial_y^2 \sigma \\ \dot{\gamma} &= \dot{\gamma}_0 - w \\ \partial_t w &= -\frac{\dot{\gamma}}{\Gamma}(w - w^*). \end{aligned} \quad (2)$$

Here we have introduced a proportionality constant, $\tau = \Gamma/\dot{\gamma}$, which can be fitted, when comparing simulations to the predictions of the model. For the stability analysis, it is irrelevant. Higher-order diffusive terms may be added to the stress and/or the w -equation. We have checked that the inclusion of such terms does not change the stability analysis. Other hydrodynamic models of granular fluids include velocity fluctuations [16], e.g. granular temperature which, however cannot explain effects due to friction.

The model allows for two stationary states: The first one corresponds to stationary flow and is explicitly given by

$$\dot{\gamma} = \dot{\gamma}_0 - w^*; \quad w = w^*; \quad \sigma = \sigma_0 \quad (3)$$

Given that w^* should vanish for vanishing shear, we take it as $w^* = b\sigma$, so that we recover the constitutive relation for frictional grains, Eq. (1). The second stationary solution accounts for the jammed state and reads

$$\dot{\gamma} = 0; \quad w = \dot{\gamma}_0; \quad \sigma = \sigma_0 \quad (4)$$

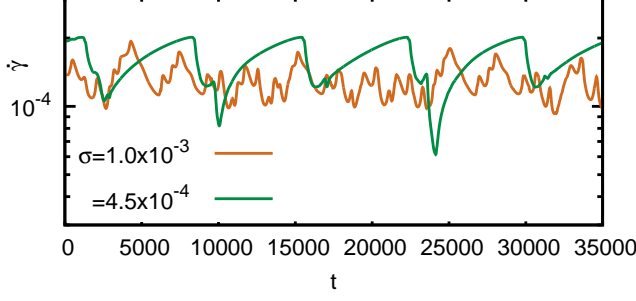


FIG. 2. Spatially averaged strain rate vs. time from numerically integrating the hydrodynamical model for different imposed stress. In the unstable region at intermediate stress-values we observe oscillations and chaotic solutions ($\Gamma = 10^{-3}$, $\phi = 0.7975$).

For both stationary states, the stress $\sigma = \sigma_0$ is homogeneous over the sample, in agreement with the Navier-Stokes equation which require a homogeneous stress in two dimensions and hence do not allow vorticity banding.

Stability analysis - In order to study the stability of the stationary states, we consider small deviations $\delta\sigma, \delta w \sim e^{\Omega t} e^{iky}$ and linearise Eq.2 in $\delta\sigma, \delta w$. As expected the stationary flow is unstable for $\frac{\partial \dot{\gamma}}{\partial \sigma} < 0$. Below ϕ_c , this does not occur and we find two stable modes: a hydrodynamic one, $\Omega_1 \propto -k^2$ corresponding to the conservation of momentum, and a nonhydrodynamic one $\Omega_2 \propto -\frac{\partial \dot{\gamma}}{\partial \sigma} \rightarrow 0$, corresponding to the relaxation of the microstructure, whose relaxation time becomes infinite as $\phi \rightarrow \phi_c$ and $\sigma \rightarrow \sigma_c$. Above ϕ_c , the model predicts two stable stationary flow solutions, inertial flow at small stress and plastic flow at large stress. In between a gap of unstable stress values occurs, such that no stationary homogeneous flow is possible in this range of stresses. A typical flow curve in the range $\phi_c < \phi < \phi_\sigma$ is shown in the inset of Fig. 3 as line 1, indicating the unstable regime as red. The jammed state is only stable for $\phi > \phi_\sigma$ in the region where the constitutive relation yields a negative $\dot{\gamma}$. A typical flow curve in the range $\phi_\sigma < \phi < \phi_\eta$ is line 2 in the inset of Fig. 3 with the stable jammed state marked in blue. For $\phi > \phi_\eta$, only stationary plastic flow and stationary jamming are predicted by linear stability analysis. The resulting phase diagram of the model is shown in the main panel of Fig. 3.

Above the critical packing fraction, ϕ_c , a finite range of inaccessible stress values with $\frac{\partial \dot{\gamma}}{\partial \sigma} \leq 0$ exists and gives rise to a corresponding range of unstable wavenumbers $k^2 \leq k_c^2 = \left| \dot{\gamma} \frac{\partial \dot{\gamma}}{\partial \sigma} \right|$, which shrinks to 0 as $\phi \rightarrow \phi_c$. Hence the model predicts an approximately harmonically oscillating state at the onset of instability, while well inside the unstable region more and more wavenumbers are unstable so that one expects a broad range of frequencies to be present in the spectrum. These expectations are born

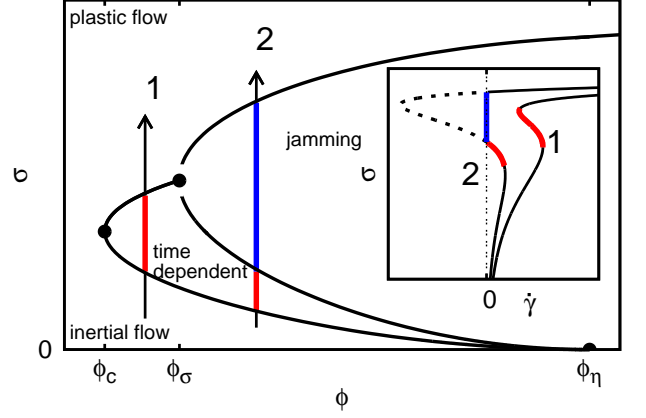


FIG. 3. Inset: Typical stationary states (flow curves) in the range $\phi_\sigma > \phi > \phi_c$ (1) and $\phi > \phi_\sigma$ (2). Unstable regions are highlighted in red (time dependent flow) and blue (jamming). Main: Phase diagram following from the linear stability analysis; the two generic flow curves, displayed in the inset, correspond to the paths, denoted by 1 and 2.

out by numerical integration of the partial differential equations in order to obtain the full nonlinear dynamical evolution. Close to ϕ_c , the oscillations are approximately harmonic, while we find oscillating and seemingly chaotic solutions at larger packing fractions (see Fig. 2).

Comparison with simulations - To check the predictions of the above analysis, we performed stress controlled simulations (for technical details see Ref. [7]) along the paths 1 and 2 in the phase diagram. The time-dependent strain rate along path 1 is shown in Fig. 4a for three different stress values. The lowest one shows stationary flow in the Bagnold regime, the chaotic time-dependence with oscillatory components is represented by the two red curves for intermediate stress and larger stress gives rise to stationary plastic flow (not shown in Fig. 4a). Similarly in Fig. 4b we show the strain rate as a function of time for path 2. In addition to the steady-state flow, and the oscillatory flow, there are stationary jammed states, where the initial flow ceases after a certain amount of time.

To get a quantitative measure of the irregularity in the time dependence of the states, we have computed the power spectrum $C(\omega)$ as the Fourier transform of the strain rate auto-correlation function (Fig. 5). In the stable regimes, the power spectrum is $C \sim \omega^{-2}$, suggesting simple exponential correlations and linear noise. In the unstable regime this background spectrum is superposed by additional and irregular complex structures. This is a strong indication of truly nonlinear chaotic dynamics. Noteworthy is also the strong peak at higher stresses. This corresponds to the fast oscillations visible in Fig. 4a.

Finite-size effects - Snapshots from video 1 indicate

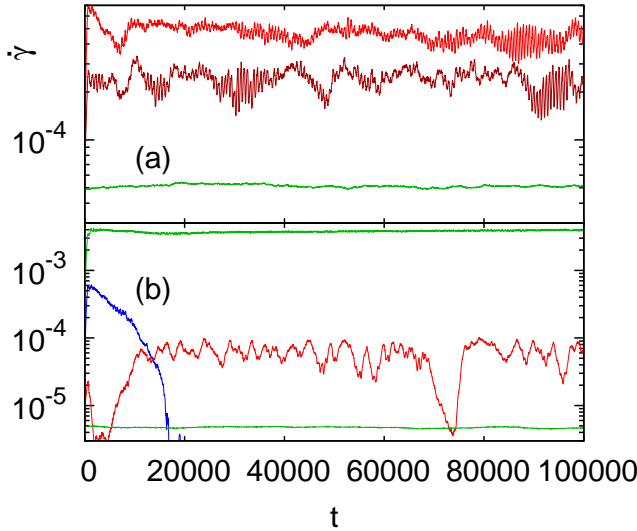


FIG. 4. Strain rate $\dot{\gamma}(t)$ as a function of time for several values of stress, corresponding to path 1 (top, $\phi = 0.7975$) and path 2 (bottom, $\phi = 0.8035$) in Fig. 3; the sudden drops of the strain rate in the time dependent flow curve indicate that the system nearly gets jammed.

the build-up of large-scale coherent structures. A large correlation length has also been observed in Ref. [3] in the context of continuous shear thickening in non-Brownian particle suspensions. These correlations are also in line with the strong finite-size effects observed in the flow curves of Fig. 1. Indeed, lowering the system size the oscillatory state acquires a finite lifetime and the system jams. This is because in small systems there is a finite probability that large strain rate fluctuations towards $\dot{\gamma} \rightarrow 0$ lead into a stable jammed state (see Fig. 4b for an example of such an excursion). Similarly, the appearance of s-shaped flow curves is due to system-size dependent strong strain rate-fluctuations towards the jammed state with $\dot{\gamma} = 0$.

Conclusion - We discuss stress-controlled driving of a granular system that undergoes discontinuous shear thickening. In particular a regime is identified where the system does not settle into a time-independent steady-state. Instead, it displays spatio-temporal oscillations and chaotic behavior as a novel possibility to adopt to stress in the unstable parts of the flow curve. Recent experiments on corn-starch reveal very similar unsteady flow where theory predicts discontinuous shear thickening [17].

Simulations reveal a phase diagram that has a characteristic re-entrant form with steady-state flow at small and large stresses. At intermediate values of stress either time-dependent states are observed or the system settles into a non-flowing jammed state, depending on stress and packing fraction. For $\phi_c < \phi < \phi_\sigma$ only time dependent solutions are observed, whereas for $\phi_\sigma < \phi$ a sequence of

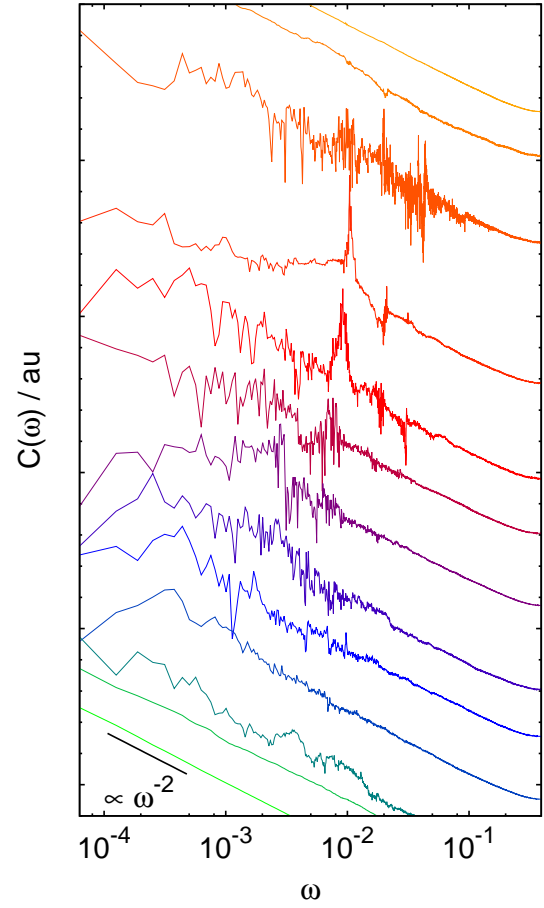


FIG. 5. Power spectrum $C(\omega)$ of strain rate fluctuations at constant stress from low (bottom) to high (top) values of σ (shifted vertically for clarity of presentation); in the stable region $C \sim \omega^{-2}$, whereas in the unstable region additional complex structures are superimposed on the ω^{-2} decay.

inertial flow, chaotic flow, jammed state and plastic flow are seen for increasing values of σ , until at $\phi = \phi_\eta$ only a transition from the jammed state to plastic flow remains.

We also present a hydrodynamical model, coupling stress to a microstructural observable. Within linear stability analysis we recover the detailed features of the phase diagram as obtained from simulations. In the unstable region, the model predicts either oscillating or time-chaotic flow.

In future work we plan to quantify the spatio-temporal correlations that are visible in the snapshots and compare them with length-scales determined in [3] from velocity correlations. We furthermore aim to better understand the microstructural observable, check whether it can be associated with the contacts which are blocked by Coulomb friction and explore the possibility of a tensorial observable, such as the fabric tensor.

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Appendix

In this appendix we deal with flow curves of frictional grains at fixed packing fraction when the particles' elastic and viscous damping constants change.

In the main article we use unit strength for elastic and viscous contribution in the linear-spring dashpots (k and η , respectively). Units of time and energy dissipation when particles interact are set by this choice. In particular, the coefficient of (normal) restitution, $\epsilon^{(n)}$, and the binary collision time, t , are set. Table I summarizes the normal coefficient of restitution and the binary collision time for the parameters that we discuss here. We tune both contributions, $k = k^{(n)} = k^{(t)}$ and $\eta = \eta^{(n)} = \eta^{(t)}$, independently. Both, $\epsilon^{(n)}$ and t , just serve as a guidance

TABLE I. Normal restitution coefficient $\epsilon^{(n)}$ and binary collision time t for different k and η .

k	η	$\epsilon^{(n)}$	t
1	1	0.305	2.375
1	1/2	0.569	2.26
1	1/10	0.895	2.22
1/2	1	0.163	3.63
2	1	0.44	1.4
10	1	0.7	0.7

on properties of pairwise collisions and do not respect the large packing fraction. Also the important frictional contribution which implies tangential restitution $\epsilon^{(t)}$ is not characterized by these quantities. The normal restitution can be computed easily while the tangential part is of rich and complicated behaviour due to its dependence on the impact velocities [20].

The following data shows flow curves of a system with $N = 8000$ particles. Fig. 6 shows scaled flow curves for fixed $\phi = 0.80$ and $\eta = 1$. When k decreases ($\epsilon^{(n)}$ decreases), the discontinuity shifts towards small strain rate until it is missed out by our simulation. Note that the numerical effort for low strain rate is much larger as for large strain rate. An increasing k (increasing $\epsilon^{(n)}$) shifts the discontinuity towards larger strain rate which makes the discontinuity smaller until it vanishes. At $k = 10$ the transition from inertial to plastic flow is smooth and without shear thickening. The phenomenology of varying k seems in that range similar to a change of the packing fraction. The latter was studied in previous work [7]. A systematic study of variations of all three parameters, ϕ , k and η , is out the scope of this article. In Fig. 7 we

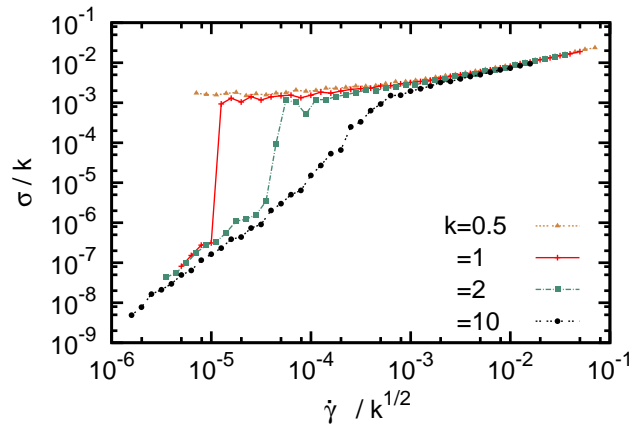


FIG. 6. Scaled flow curves for $\phi = 0.80$, viscous damping parameter $\eta = 1$ and varying elastic constant k .

show a choice of packing fractions with phenomenology similar to Fig. 6. Lower ϕ leads to flow curves similar to

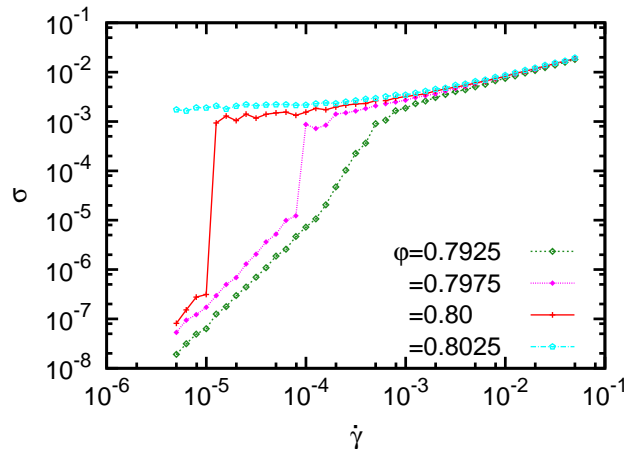


FIG. 7. Flow curves for varying packing fraction ϕ across the transition with viscous damping parameter $\eta = 1$ and elastic constant $k = 1$.

those with large $\epsilon^{(n)}$ and large ϕ show the same behaviour as small $\epsilon^{(n)}$. The variation of the viscous damping parameter η is shown in Fig. 8. The phenomenology is the same as above: decreasing $\epsilon^{(n)}$ (larger η) leads to a shift towards small strain rates and an increasing coefficient of restitution (smaller η) shifts the discontinuity towards larger strain rate until it vanishes. Then the transition between inertial and plastic flow is smooth and without shear thickening again.

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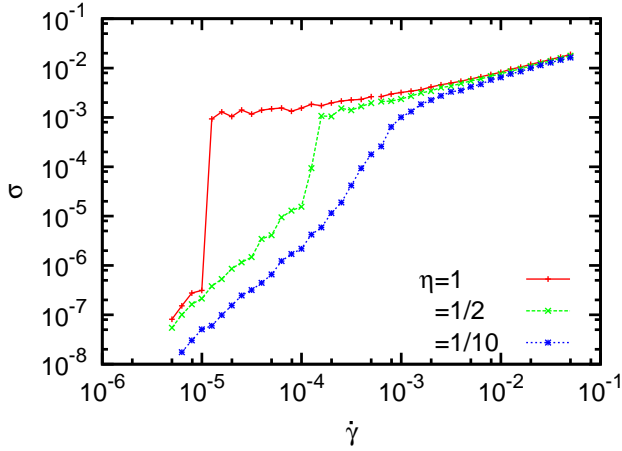


FIG. 8. Flow curves for $\phi = 0.80$, elastic constant $k = 1$ and varying viscous damping parameter η .

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